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FTD-TT- 62-1114/1+2

UNEDITED ROUGH DRAFT TRANSLATION

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English Pages: 15

SOURCE: Akademiya Nauk SSSR. Fizicheskiy Institut Imeni P. N. Lebedev, Issledovaniya po Eksparimentalnoy i Teoreticheskoy Fiziki, Sbornik, Moskva, 1959, pp. 62-70

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FTD-TT- 62-1114/1+2 Date 11 Sept 1962

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As is known, more than forty years ago A. Einstein [1] made his presentation about the phenomena of negative absorption (induced emission). The existence of negative absorption was not questioned by anyone because it was an unavoidable consequence from very general considerations. However, the matter of direct experimental confirmation of the existence of negative absorption stood in much worse circumstances. The investigations of Ladenburg [2], independently of an evaluation of their demonstrability, were devoted to the effect of negative dispersion, and not to absorption. These two effects, of course, are inseparably connected, but just the same they are different in their nature. In this sense the experimental checking of the formula of Planck, centainly, represented a more direct way of confirming the existence of the phenomena of negative absorption (term in the denomenator of Planck's formula)[1].

The difficulty in observing negative absorption is to be found in the fact that under conditions of equilibrium negative absorption overlaps with ordinary absorption.

In 1940 one of the authors of this article [3] indicated the conditions in which one should expect negative absorption to prevail over ordinary absorption, and there was formulated the principle of molecular amplification: "For this it is necessary that $\frac{N_k}{N_j}$ exceed the ratio of static weights $\frac{q_k}{q_j}$. The lat-

[1] It is interesting to note that negative absorption assures the transition from Planck's formula to the Rayleigh-Jeans formula in the field of low frequencies. This indicates a more classical nature for negative absorption than is generally accepted.

ter has not been once observed in a discharge notwithstanding the fact that such a relationship of concentration can in principle be realized ... In such experiments we will get an intensity of outgoing radiation greater than the incident one, and it would be possible to speak about a direct experimental demonstration of the existence of negative absorption" [3].

The indicated conditions were first realized in certain experiments by Lamb in measuring the displacement of the hydrogen levels [4], where negative absorption is used for breaking down the metastable atoms of hydrogen.

Basov and Prokhorov, as well as Gordon, Zeiger, and Townes very delicately made use of the above-indicated principle of radiation amplification for creating a molecular generator of microwave radiation [5].

Basov and Prokhorov gave a detailed theory for the molecular generator [5].

The aim of the present experimentation consisted in the investigation of the effect of negative absorption in the ordinary optical range. Preliminarily an analysis is made of the peculiarity of the optical properties of the medium with a negative factor of absorption[1].

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[1] In the patent clain by V. A. Fabrikant, M. M. Vudynskiy, and F. A. Butayeva of June 18, 1951, and in the supplement to it of June 16, 1951 (priority statement No. 0270--2423 MPSS) there was a brief exposition of the theory of optical amplifications of radiation (now called "masers") and the possibility was indicated of the application of the given principle for the amplification of radio waves. In the same text there was described a method for obtaining inequilibrized conditions by irradiating the medium from an auxiliary source of waves, which, as is known, has found practical application in "masers (Note of the authors in correcting).

Obtaining a Negative Absorption Factor

We recall some known relationships. The absorption factor k_ν is connected

with the —— probabilities of absorption B_{12} and the negative absorption B_{21} in the following form

$$\int k_\nu d\nu = \frac{h\nu}{c}(B_{12}N_1 - B_{21}N_2), \tag{1}$$

where the integral is taken with the limits of the spectral lines corresponding to the transition $2 \rightarrow 1$, N_1 and N_2, the concentration of the atoms or molecules at the levels E_1 and E_2.

In conditions of thermodynamic equilibrium

$$(A_{21} + B_{21}\rho_\nu)N_2 = B_{12}\rho_\nu N_1, \tag{2}$$

where A_{21} is the probability of spontaneous

hence

$$(B_{12}N_1 - B_{21}N_2) = \frac{A_{21}N_2}{\rho_\nu}, \tag{3}$$

i. e.,

$$(B_{12}N_1 - B_{21}N_2) > 0; \int k_\nu d\nu > 0 \tag{4}$$

and, consequently, the phenomena of ordinary absorption prevails over the phenomena of negative absorption. This is — connected with the equilibrised character of the distribution of atoms over energy levels.

By making use of the known connection between B_{12} and B_{21} we get:

$$\int k_\nu d\nu = \frac{h\nu}{c} B_{12}N_1\left(1 - \frac{g_1}{g_2}\frac{N_2}{N_1}\right). \tag{5}$$

In equilibrised conditions

$$\frac{N_2}{N_1} = \frac{g_2}{g_1} e^{-\frac{E_2 - E_1}{kT}} \tag{6}$$

and the second member in parenthesis is less than unity.

In this way under conditions of thermodynamic equilibrium the absorption factor is always positive.

Only under conditions of nonequilibrium one can expect to get a medium with a negative —— absorption factor because in these conditions equality (2) is no longer valid. From (5) it is — clear that —— for obtaining of the negative factor of absorption it is necessary to accomplish the inequality

$$\frac{N_2}{N_1} > \frac{g_2}{g_1}. \tag{7}$$

Within the limits with very large $\frac{N_2}{N_1}$

$$\int k_\nu d\nu = -\frac{h\nu}{c} B_{21} N_2. \tag{8}$$

Inequilibrized conditions can be both stationary and nonstationary. In stationary conditions the ratio of concentrations is determined by the simple equality

$$\frac{N_2}{N_1} = \frac{\alpha_2 \tau_2}{\alpha_1 \tau_1}, \tag{9}$$

where α_1 and α_2 are the numbers of the cases of excitation of atoms in one second to the levels E_1 and E_2 and τ_1 and τ_2 are the durations of the lives of the atoms at these levels.

The number of cases of excitation includes both the optical nonoptical processes.

The τ is also expressed as

$$\tau = \frac{1}{\Sigma W_i}, \tag{9a}$$

where W_i represents the probabilities of destruction of the excited atoms through possible processes.

The inequality (7) is converted into

$$\frac{\alpha_2 \tau_2}{\alpha_1 \tau_1} > \frac{g_2}{g_1}. \tag{10}$$

The inequality —— (10) makes it possible to select conditions favorable for getting a negative absorption factor.

For example, it is clear, that the selective abridgement of the durability of the life of the low level represents one of the ways for getting the necessary conditions. Another way is the selective excitation up to the high level.

In the practical accomplishment of the experiments the value of the absorption factor is also substantial, and this is determined by the probability B_{21}, the concentration of atoms N_2 and the frequency ν. In particular, if the upper level is metastable the concentration of atoms at it usually is higher than at a lower level which has the same life durability, but in this case it is important that it lead to little probability of optical transition of A_{21} to a lower level.

As is known [6]

$$A_{21} = \frac{64\pi^4 e^2 \nu^3}{3hc^3} |r_{21}|^2, \tag{11}$$

Hence

$$B_{21} = \frac{8\pi^3 e^2}{3h^2} |r_{21}|^2. \tag{12}$$

If the smallness of A_{21} is connected with the smallness of the square of the dipole moment $[\bar{r}_{21}]^2$, — then to the low A_{21} there will correspond the low B_{21}, and observation of the effects of negative absorption become much more difficult.

If, on the other hand, the smallness of A_{21} is connected with the smallness of ν, and the dipole moment is great, then favorable conditions exist for observing the effects brought about by negative absorption. Just such a situation occurs in the microwave range. On the other hand, despite the great value for B_{21}, the conditions of the experiments by Lamb and of the microwave generator correspond to small absolute values for the absorption factor. This is explained, according to (8), by the low frequency of ν and small concentration of N_2.

In the experiments by R. Ladenburg both the frequency ν and the concentration N_2 have much greater values (visible part of the spectrum and discharge in inert gases), but the inequality — (10) is not fulfilled. As is known, in the usual electrical discharge the concentration of atoms at the upper levels is less and not greater than the Boltzmann ones [3]. Therefore the cases of negative absorption not only do not exceed these of ordinary absorption, but their role is less — even than in equlibrium conditions.

In the optical range often

$$\tau_2 \simeq \frac{1}{A_{21}}, \tag{12a}$$

Then, according to (8) and (9)

$$\int k_\nu d\nu = -\frac{\lambda^2}{8\pi} a_2. \tag{13}$$

By taking the length of — wave $5 \cdot 10^{-5}$, width of line of the order 10^8—10^9 cm^{-1}, and k_ν of the order 10^{-3} cm^{-1} for a_2 we obtain a value of the order

10^{15}—10^{16}, which corresponds to a supply of power of 10^{-4}—10^{-3} W/cm^3.

We undertook to create conditions corresponding to the great absolute values of the absorption coefficients. Before passing on to these experiments one should consider the peculiarities of the optical properties of the medium with negative absorption factor.

Optical Properties of the Medium with Negative Absorption Factor

As is known, radiation occurring as a result of the phenomena of negative absorption should be consistent with the radiation causing the phenomena and have the same direction. The radiation of a medium brought about by spontaneous emission, generally speaking, is inconsistant with this radiation. For the coherent part of the intensity of the beam being propagated in a medium with negative absorption factor one can write an equation:

$$\frac{dI_\nu}{dx} = |k_\nu| I_\nu \qquad (14)$$

Hence

$$I_\nu(x) = I_\nu(0) e^{\int^x |k_\nu| dx}. \qquad (15)$$

The observable intensity will be greater than $I_\nu(x)$ on account of the spontaneous radiation of the medium, but when working with small solid angles one may make the role of the spontaneous radiation small. In the molecular generator the directability of the negative absorption, apparently, is not so important.

To the equation (14) there corresponds the avalanche-like increase in the intensity in proportion to the propagation of the beam, which is analogous to the electron avalanche in a discharge.

One should remember that the absorption factor k can depend very much on I_ν. Instances of negative absorption can noticeably decrease the durability of the life of τ_2 atoms at the upper level transferring them to the lower level (which at the same time increases τ_1). In the experiments of Lamb τ_2

is simply inversely proportional to the intensity of the radiation, so that the metastable atoms are broken down practically only on account of the negative absorption.

Besides, in these conditions —— the formula (8) is valid because $N_2 > N_1$. Therefore the absorption factor k is inversely proportional to I_ν, and the equation (14) takes the form

$$\frac{dI_\nu}{dx} = a, \tag{14a}$$

where is the constant

Then instead of (15) we get

$$I_\nu(x) = I_\nu(0) + ax. \tag{15a}$$

The correlation (15a) is valid only for sufficiently great intensities where τ_2 is determined by the instances of negative absorption.

From (15a) it follows that $I_\nu(x)$ is not proportional to $I_\nu(0)$, i. e., we have a case of nonlinear optics.

Generally with k_ν depending on I_ν nonlinear effects occur.

The properties of the medium depend on the intensity of the radiation passing through it, and thus there is broken down the principle of superposition (analogous questions for positive absorption factor are considered by S. I. Vavilov [7]). The indicated circumstance sets a limit to the avalanchelike increase in intensity and restricts the area of the applicability of the relationship (15). The maximum attainable value for the absorption factor is proportional to α_2, which characterizes the excitation of the atoms up to the upper level. In this case there occurs practical equality between α_2 and the number of the instances of negative absorption in one second.

Due to the negative absorption all the energy supplied to the atoms in excitation is radiated within the limits of a small solid angle, determinable by the angular opening of the beam. Basov and Prokhorov [5] pointed to the nonlinearity as the factor determining the amplitude of the oscillations in the

molecular generator.

The transparence of the layer of the homogeneous medium L_ν under conditions of the validity of the relationship (15) is equal to

$$T_\nu = e^{|k_\nu| L}, \tag{16}$$

where L is the thickness of the layer.

In this way the transparency of the layer exceeds unity, which does not contradict, of course, the law of conservation of energy. By virtue of the negative absorption there occurs a redistribution along the angles of the energy of radiation emitted by the excited atoms of the medium, and the probability of the optical processes of emission of energy is increased.

Let us pass on to a consideration of the radiation proper of the homogeneous plane layer of a medium with a negative absorption factor. In assuming the layer to be homogeneous we disregard the nonhomogeneity brought about by the events of negative absorption, i. e., we limit ourselves to the field of the applicability of the relationship (16).

From a layer of the thickness dx outside there is emitted a radiation of the intensity

$$dI_\nu = j_\nu e^{|k_\nu| x} dx, \tag{17}$$

where j_ν is the emission capacity of the medium.

From (16) it follows that

$$I_\nu = \frac{j_\nu}{|k_\nu|}\left(e^{|k_\nu| L} - 1\right). \tag{18}$$

Since the conditions are not in equilibrium, the ratio $\frac{j_\nu}{k_\nu}$ cannot be equated with the emission capacity of an absolutely black body. It is generally clear that for such conditions Kirhhoff's law is altogether inapplicable.
In a medium with a positive absorption factor with the increase in the thickness of the layer the conditions approach equilibrium, and the intensities of the spectral lines, as a rule, smooth out. In a medium with negative absorption factor, on the other hand, in accordance with (18), with the increase in the thickness there can be an increase in the intensities of the spectral

lines. This is connected with the fact that greater k generally corresponds to greater j_{ν} (proportionality between A_{21} and B_{21}). Besides, it follows from (18) that in proportion as the thickness of the layer increases, the half width of the spectral lines should not increase but decrease, since the greater k_{ν} corresponds to the center of the spectral line. Attention was also focused on this circumstance in the theory of the molecular generator [5].

In accordance with (18) the indicatrix of radiation of the layer will be broader than the Lommel one, corresponding to k_{ν}, equal to zero.

The problem of the limiting intensities cannot be analyzed on the basis of the correlation (18), since with this we pass outside of the limit of the area of the applicability of this relationship. In any case with a negative there will not occur a rapid saturation of I_{ν} with the increase of L, which is characteristic for media with a positive absorption factor.

Experiments in the Creation of a Medium with a Negative Absorption Factor

Since in the visible part of the spectrum the frequencies are of the order of 10^{15} sec^{-1}, to the forbidden spontaneous transitions there should correspond infinitesimally small dipole moments. Therefore the experiments in this region of the spectrum should be based on the use of allowed transitions, which makes difficult the obtaining of the necessary ratio of concentrations of excited atoms at the upper and lower levels. The spontaneous transitions reduce τ_2, increase n_1, and thereby obstruct the obtaining of the conditions corresponding to the inequality (10). In this sense the microwave range is much more favorable. In this connection high frequency is favorable from the point of view of the absolute value of the absorption factor [equation (8)].

Our first trial was based on the method in which the atoms are indirectly excited up to the M upper level and get to the lower level only through

spontaneous and forced — transitions from the upper level. This proves to be analogous to the "sorting out" of the molecules in the molecular generator (5). Apparently, such a situation gives the most favorable ratios — of τ_1 and α_2 for a given pair of excited levels. For getting ——— suitable ratios of τ_1 and τ_2 it is important that from the top level only lines begin used for observation of negative absorption.

The presence of "excess" transitions leads to undesirable shortening of τ_2.

One could expect that corresponding conditions occur in the fluorescence of the vapors of cesium produced by the intensive helium line 3,888.65. As was established by Boecner [8], this line gives rise to the transition $6^2S_{1/2} \rightarrow 8^2P_{1/2}$, to which in the spectrum of cesium there corresponds the line 3,888.65. With the addition of some inert gas (for example, the same helium), the atoms of cesium because of the collisions pass from the level $8^2P_{1/2}$ to the level $8^2D_{3/2, 5/2}$ (distance of 0.01 ev).

In the spectrum of fluorescence there appear the lines 6,983($8^2D_{3/2} \rightarrow 7^2P_{1/2}$), 6,723($8^2D_{3/2} \rightarrow 7^2P_{1/2}$), and 6,973($8^2D_{5/2} \rightarrow 7^2P_{3/2}$).

The durations of life of the upper levels 8^2D—τ_2 should be more than τ_1 for the lower levels 7^2P.

This is explained by the fact that the probabilities of spontaneous transitions ——— $8^2P \rightarrow 7^2P$ should be considerably less than the probabilities for the transitions $7^2D \rightarrow 6^2S$ corresponding to the resonance line of cesium. A complicating circumstance is afforded by the "capture" of the resonance radiation raising the effective duration of the life of the atoms at the levels 7^2P. Unfortunately for cesium data are lacking needed for quantitative evaluation ratio τ_1 to τ_2. Nevertheless, on the whole one should expect the fulfillment of the inequality (10). The vapors of cesium filled a vessel of the Wood type with a length of 200 mm and diameter of 28 mm. Into the vessel there were added helium and other inert gases with pressures of the order of some

few mm of the Hg column. The fluorescence of the ———— vapors was excited with the aid of three loop-shaped helium lamps located on the sides of the vessel. The basic part of the vessel and the lamps were enclosed in a reflecting jacket heated from the outside. The pressure of the vapors of cesium was regulated with aid of an extension kept at a lower temperature than the basic furnace.

The diameter of the helium lamps was 10 mm, length 300 mm, current strength 200--500 ma, and voltage 5,000 v.

The slit of the three-prism spectograph of the Zeiss Company was located in the main focus of the condenser lense, the optical axis of which coincided with the axis of the vessel with vapors.

For the lines 6,983, 6723, and — 6,973 — the probabilities of B_{21} have ratios, according to the rule of sums, as 1 : 5 : 9. Therefore, on the basis of (18) one should expect changes in the ratios of the intensities of these lines with changes in the intensities of the exciting radiation.

Particularly, with an increase in the intensity of the exciting radiation the intensities of the lines 6,723 and 6,973 should increase with relation to the line 6,983.

Actually with the temperature of the extension of the resonance vessel at 120°C corresponding effects were obtained. In the table there are compiled the results for two values of the currents in the helium lamps.

Notwithstanding the obtaining of effects with the necessary sign, the method in question cannot be considered sufficiently direct. Besides, the too large value for the observed effect arouses doubt. Checking experiments with change in the length of the radiating layer did not give clear results. Therefore another more direct method was used.

As a second method for obtaining a negative factor of absorption there was used an artificial shortening of the life duration of the excited atoms

at the low levels

Table

Ratios of Intensities

Intensity	Current, ma 200	500
$\frac{I_{6923}}{I_{6983}}$	3.9	5.3
$\frac{I_{6973}}{I_{9983}}$	5.4	6

As a subject for investigations there was chosen the visible triplet of mercury $7^3S_1 \rightarrow 6^3P_{0,1,2}$.

The vapors of the mercury were excited with the aid of a glow discharge. In the cathod parts of the glow discharge there are comparatively fast electrons, which assures a favorable ratio for α_1 and α_2. For shortening the life duration of the lower levels there was added hydrogen with a pressure of some mm of the Hg column.

As is known hydrogen powerfully breaks down the excited atoms of mercury at the levels 6^3P. With the aid of a narrow mercury lamp there was determined the transparency of the discharge tube with a mixture of vapors of mercury and hydrogen. With this objective using two lenses, the image of the slit illuminated by a narrow lamp was focused on the input slit of a monochromator. Between the lenses in a parallel space there is located a discharge tube. The length of the tube is 360 mm (L) and the diameter 75 mm. Behind the output slit of the monochromator there was a photo-multiplier connected with a sensitive galvanometer.

With the aid of the described setup there were observed right along values for transparency above unity, i. e., the effect of negative absorption in the pure form.

As an illustration we present data obtained with a pressure of the mer-

cury vapors of the order of 10^{-3} mm of the Hg column, pressure of the hydrogen of 0.3 mm of the Hg column, and a discharge current of 70 ma

λ	5461	4358	4047
T_{λ}	1.10--1.14	1.08--1.10	0.95--1.0

The experiments were repeated with the addition of helium with a pressure of 4 mm of the Hg colum to the mercury vapors. The current of the discharge was 125 ma. The addition of helium unexpectedly lead to the obtaining of even greater effects.

λ	5461	4358	4047
T_{λ}	1.14--1.29	1.13--1.23	1.20 (?)

Decreasing the pressure of the helium led to a decreasing of the effects. For a check measurements were made in the direction perpendicular to the initial one when there was a sharp diminution in the length of L. These measurements gave a sharp diminution of the effects. It is interesting to note that the cooling of the discharge tube (lowering the pressure of the mercury vapors) led to a change of the sign of the absorption effects, i. e., the transparency for all the lines became less than unity. In this case, as usual, absorption has greater values for the lines 5461 and 4047 Å, which end at the metastable levels and less for the line 4358 Å.

The theoretical interpretation of the results obtained is made more difficult by the fact that there are no dependable data on the energies and concentrations of electrons in a discharge of this type.

The values obtained for the transparency in accordance with (16) correspong to k_0, approximately equal to 0.005. If one considers the form of the lines as of the Doppler type and makes use of the formula (8) (A_{21} of the order 10^{+8} sec^{-1}), then for the concentration of the atoms at the top level N_2 we get a value of the order of 10^9. For the given conditions this value is high. On the other hand attempts to explain the values obtained for transpar-